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(71)Applicant : MATSUSHITA ELECTRIC IND CO LTD

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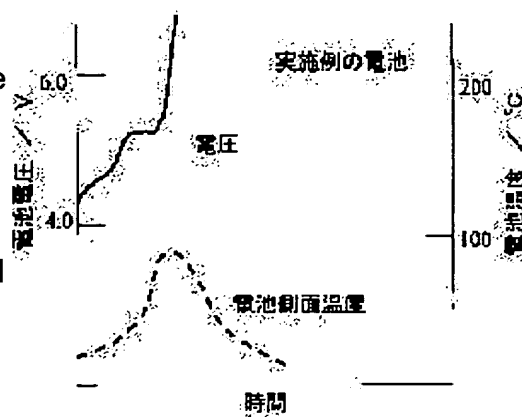
(72)Inventor : WATANABE SHOICHIRO
IWAMOTO KAZUYA
UEDA ATSUSHI
NUNOME JUN
KOSHINA HIDE

(54) NON-AQUEOUS ELECTROLYTE SECONDARY BATTERY SYSTEM CONTAINING NON-AQUEOUS ELECTROLYTE SECONDARY BATTERY AND CHARGE CONTROL SYSTEM AND APPARATUS MOUNTING THIS

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a battery having excellent safety and long time reliability and a portable information apparatus.

SOLUTION: In the rechargeable non-aqueous electrolyte secondary battery that uses a positive electrode, a negative electrode and a non-aqueous electrolyte, an organic compound that has -8.5 eV--11.0 eV of HOMO (highest occupied molecular orbital energy) as measured using PM3 method to Hamiltonian in the semi-empirical molecular orbital measuring method and -0.135 eV-3.5 eV of LUMO(lowest unoccupied molecular orbital energy), preferably, phenylcyclohexane is added 0.1-20 wt.% to the total of the electrolyte additive and the non-aqueous solvent, and this battery and a charge control system are composed.



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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the charge control system of a nonaqueous electrolyte rechargeable battery and said cell.

[0002]

[Description of the Prior Art] In recent years, portable-izing of electronic equipment, such as an AV equipment or a personal computer, and cordless-ization are progressing quickly, and small and the demand to the rechargeable battery which is lightweight and has a high energy consistency are high as these power sources for a drive. The nonaqueous electrolyte rechargeable battery using the negative electrode which uses a lithium as an active material in this has great expectation as a cell which especially has a high voltage and a high energy consistency.

[0003] By the above-mentioned cell, the lithium content metallic oxide which shows a 4V class electrical potential difference to positive active material is used, and the ingredient which can intercalate and deintercalate lithiums, such as for example, a carbonaceous ingredient, is used for a negative electrode.

[0004] In such a nonaqueous electrolyte cell, reservation of safety is one of the most important technical problems.

[0005] Especially, when it charges in a rechargeable lithium-ion battery exceeding a predetermined charge electrical potential difference by failure of a charge control circuit etc., for example, it will be in a overcharge condition, and the lithium ion of a positive electrode will be drawn out superfluously, it will move to a negative electrode, and the lithium more than predetermined design capacity will deposit as a metal lithium on occlusion or a negative-electrode front face in a negative electrode. When charge is continued still more compulsorily in such the condition, the internal resistance of a cell rises and generation of heat by the Joule's heat becomes very large, and abnormality generation of heat and when the worst, it may result in a thermal run away. Generally a means to sense internal pressure change of a cell with being indicated in order to solve such a technical problem (U.S. Pat. No. 4943497 specification), and to intercept the charging current etc. is used.

[0006] However, by such mechanical current cutoff device, the top where a cost cut is difficult, it is becoming [a cell] difficult small and to insert in the interior of a cell structurally as it thin-shape-izes.

[0007] The additive which causes a reversible oxidation reduction reaction to the electrolytic solution is added in a cell to such a technical problem, and the approach of carrying out self-consumption of the electrical energy switched on in the cell as a redox shuttle is proposed. (For example, JP,1-206571,A, JP,6-338347,A, JP,7-302614,A, etc.) .

[0008] However, by the approach using such a redox shuttle, when a overcharge current becomes large, since a limitation is in a charge transfer reaction rate and the passing speed of a lithium ion, it cannot be said that it is safe enough.

[0009] As opposed to such a technical problem, by JP,9-50822,A and JP,10-50342,A, the aromatic compound which has a methoxy group and a halogen radical in a cell is added, and when these additives

carry out a polymerization at the time of overcharge, a means to cause a temperature rise and to secure safety is proposed.

[0010] Moreover, or it raises the internal pressure of a cell from which a biphenyl and a thiophene are added to the electrolytic solution, and these additives carry out a polymerization and raise the electrical potential difference of a cell in JP,9-106835,A and JP,10-321258,A at the time of overcharge, a conductive polymer is constituted and the method of preventing the thermal run away at the time of overcharge is proposed by making the pass which can carry out an electronic transition in a cell generate.

[0011]

[Problem(s) to be Solved by the Invention] Although the result whose safety at the time of overcharge improves was obtained when the above additives were used, it turned out that the cycle property which is the essential property of a cell, a preservation property, etc. fall remarkably.

[0012] As a result of analyzing the degradation factor of a cell by disassembling and observing the cell after such degradation, reduction decomposition of these additives was carried out on the negative-electrode front face, the decomposition product covered the negative-electrode front face, and it became clear to degrade the discharge property.

[0013] This invention solves such a technical problem, and it offers the outstanding cell by which the safety at the time of overcharge was secured, maintaining the cycle property and preservation property which are the property of a cell essentially needed good.

[0014]

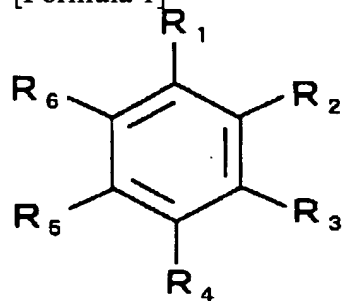
[Means for Solving the Problem] In order that this invention may solve this problem, a lithium content multiple oxide is used as positive active material. Use as a negative electrode the occlusion of a lithium, and the ingredient which can be emitted, and the nonaqueous electrolyte containing a non-aqueous solvent is used. The HOMO energy (the highest occupancy orbital energy) which used and calculated PM3 law with semiempirical molecular orbital calculus in said nonaqueous electrolyte at HAMIRUTONIAN is -8.5eV - -11.0eV. And LUMO energy (lowest unoccupied molecular orbital energy) contains the organic compound which is -0.135eV - 3.5eV as an electrolytic-solution additive.

[0015] It is desirable to add in 0.1 - 20% of the weight of the range to the sum total of a non-aqueous solvent and an additive as an amount of an additive.

[0016] As such an additive, there is an aromatic compound which is expressed with (** 1).

[0017]

[Formula 1]



[0018] (The substituent which fulfills R1-R6:H or the following conditions (the number of H is five or less pieces))

Alkyl group (you may also include a double bond)

A ring may be formed by the 1 or 2nd place (a hetero atom may also be included in a ring).

Aryl group cyclohexane (a hetero atom may also be included in a ring)

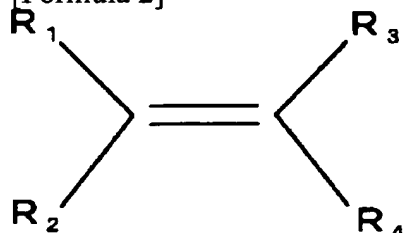
Amino group (you may have an aryl group)

For example, as an example of such an aromatic compound, hexa phenylbenzene, phenylcyclohexane, 1 and 3, 5-triphenyl benzene, para terphenyl, dodeca hydronalium triphenylene, 1-phenyl piperazine, a divinylbenzene, dicyclohexyl benzene, etc. are mentioned.

[0019] Moreover, there is unsaturated fatty acid expressed with (** 2) as an additive in addition to this.

[0020]

[Formula 2]



[0021] (The substituent which fulfills R1-R4:H or the following conditions (the number of H is three or less pieces))

Alkyl group (you may also include a double bond)

Alkyl group (you may also include an ester bond)

Alkyl group (an alicyclic compound may also be included)

Alkoxy group aryl group (a ring may be formed by R1, R2, or R3 and R4)

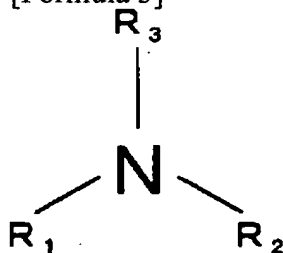
Cyclohexane (a hetero atom may also be included in a ring)

As such an unsaturated fatty acid compound, a vinyl cyclohexane, t-butyl vinyl ether, a methyl-methacrylate monomer, a cis-stilbene, a methylene cyclohexane, tetra-phenylethylene, and 2,3-dimethyl-1,3-butadiene are mentioned.

[0022] Moreover, amines which are expressed with (** 3) as an additive are sufficient.

[0023]

[Formula 3]



[0024] (Substituent alkyl group arylated alkyl radical aryl group naphthyl group which fulfills the conditions below R1 - R3:)

Tribenzylamine and N-phenyl-dibenzylamine are mentioned as such an amines compound.

[0025] Moreover, the straight chain mold organic compound which has a double bond as an additive is sufficient.

[0026] Squalene or (E)-beta-farnesene is mentioned as such an additive.

[0027] moreover -- in addition, a tris (4-methoxypheny) phosphine, a (1R)-(+)-alpha pinene, a dicyclopentadiene, the II (ethylene glycol) divinyl ether, 9, 10-dihydroanthracene, TORIPUCHISEN, [2, 2] PARASHI clo fan, etc. are mentioned as a concrete example as an additive.

[0028] A positive electrode and a negative electrode consist of the fine porosity polyolefine film or the insulating electrolytic solution, and the insulating polymer which carry out sinking-in maintenance of the electrolytic solution, and this invention functions more effectively in the nonaqueous electrolyte cell which has countered through the electrolyte which at least the part gelled.

[0029] Moreover, dependability can be raised more by using together with the charge control system of the nonaqueous electrolyte rechargeable battery which gave the function to sense the rise of cell temperature and to cut the circuit of charge.

[0030] As an approach of sensing the rise of cell temperature and cutting the circuit of charge, a positive thermistor (PTC) or a thermal fuse is desirable, and the highest dependability is acquired when the range of the operating temperature of these positive thermistors (PTC) or a thermal fuse is 60-120 degrees C.

[0031] By carrying the control system which charges such a nonaqueous electrolyte rechargeable battery and a nonaqueous electrolyte rechargeable battery, the device which is more excellent in a long-term life, and has high dependability and high safety is realizable. .

[0032]

[Embodiment of the Invention] In this invention, the organic compound contained in nonaqueous electrolyte starts electrolytic polymerization on positive active material, when a cell becomes a overcharge field. By this reaction, the reaction effectiveness by which a lithium ion is drawn out from a positive electrode can fall remarkably, and can prevent the fall of the thermal stability of positive active material.

[0033] Moreover, since the polymer generated by the polymerization is generated on a positive-electrode front face, it can check the charge transfer reaction of positive active material, it can increase the internal resistance of a cell extremely, and can stop the current from a power source.

[0034] Moreover, it becomes possible to stop a overcharge current by the manifestation of the shutdown device of the separator which originates in thermofusion, for example in the cell using the polyolefine film of fine porosity as a separator.

[0035] In this case, without a cell carrying out abnormality generation of heat, since cell temperature rises where the reaction effectiveness by which a lithium ion is drawn out from a positive electrode by adding the additive fell remarkably and the thermal stability of positive active material is held, when a current stops, cell temperature can fall gradually and can secure safety.

[0036] Moreover, by preparing the current isolating switch (for example, a positive thermistor (PTC) and a thermal fuse) of a temperature sensing mold in the cell exterior, a current can be intercepted more exactly and safety can be secured.

[0037] Since the additive in this invention has not aimed at the redox shuttle, the irreversible thing of oxidation reaction is desirable and, as for JP,7-362614,A aiming at the reversibility of an oxidation reduction reaction, and JP,9-50822,A, the purposes differ.

[0038] The organic compound added as an essential function of this invention is a potential field (in 3.0-4.3V, it must be chemically stable, and it is necessary to carry out an oxidation polymerization promptly in a overcharge field in the case where used LiCoO₂ for the positive electrode and a carbonaceous ingredient is used for a negative electrode) where a cell usually operates.

[0039] Such an electrochemical oxidation reaction can calculate PM3 law by using at HAMIRUTONIAN the orbital energy level (HOMO energy (the highest occupancy orbital energy)) of the occupancy electron which is the easiest to remove with semiempirical molecular orbital calculus. (:, for example, JP,6-333576,A, easily calculable by computer using MOPAC recently)

When HOMO energy was -8.5eV - -11.0eV, in the potential field in which a cell is usually used, it was stable, and as a result of investigating electrochemical oxidation resistance and the relation of HOMO energy, when it was overcharged, it turned out that oxidation electrolytic polymerization is carried out efficiently. Therefore, as for the organic compound as an additive in this invention, it is desirable for HOMO energy to be -8.5eV - -11.0eV.

[0040] Moreover, especially, when it was an aromatic compound and the compound which has a vinyl group as such an organic compound with which an oxidation polymerization tends to happen electrochemically, it turned out that oxidation electrolytic polymerization advances [conversion efficiency] effectively highly.

[0041] Moreover, the organic compound added in this invention needs a stable thing in the potential field (it is 0V-1.5V to Li metal-electrode criteria) of the negative electrode usually used.

[0042] It is known that the reducibility-proof of the organic compound added has the LUMO energy (lowest unoccupied molecular orbital energy) and the correlation of a molecular orbital. (For example, JP,5-290882,A) .

[0043] That is, it means that reduction potential is so low that LUMO energy is high, that is, it is hard to be returned. Such LUMO energy can use and calculate PM3 law to HAMIRUTONIAN with semiempirical molecular orbital calculus similarly.

[0044] When for example, a carbonaceous ingredient, Sn, or Si compound is used as a negative

electrode, it is desirable for LUMO energy (lowest unoccupied molecular orbital energy) to be -0.135eV - 3.5eV at least, and it is more desirable in it being 0.3-3.5eV especially.

[0045] That is, by containing such an organic compound, even if it changed into the overcharge condition, it was safe and the nonaqueous electrolyte rechargeable battery excellent in the cycle property and the preservation property was realized.

[0046] Furthermore, a more reliable rechargeable battery system can be offered by using to be a positive thermistor (PTC) or a thermal fuse as a function to sense the rise of cell temperature on the above-mentioned cell, and to cut the circuit of charge as the charge control system of the nonaqueous electrolyte rechargeable battery which it had.

[0047] As a desirable lithium content multiple oxide used for positive active material by this invention Li_xCoO_2 , Li_xNiO_2 (U.S. Pat. No. 4302518), Li_xMnO_2 , $\text{Li}_x\text{Co}_y\text{Ni}_{1-y}\text{O}_2$ (JP,63-299056,A), $\text{Li}_x\text{Co}_y\text{V}_{1-f}\text{O}_z$, $\text{Li}_x\text{Ni}_{1-y}\text{MyO}_2$ (M=Ti) V, Mn, Fe, $\text{Li}_x\text{Co}_a\text{Ni}_b\text{McO}_2$ (M=Ti, Mn, aluminum, Mg, Fe, Zr), $\text{Li}_x\text{Mn}_2\text{O}_4$, $\text{Li}_x\text{Mn}_{2-y}\text{MyO}_4$ (M=Na, Mg, Sc, Y, Fe, Co, nickel, Ti, Zr, Cu, Zn, aluminum, Pb, Sb)

(-- here -- $x = 0 - 1.2$ -- $y = 0 - 1$ -- $z = 0.9 - 1$ -- $f = 0 - 1$ -- $a + b + c = 1$ -- $a = 0 - 1$ -- $b = 0 - 1$ -- $c = 0 - 1$ -- raising -- having). Here, the x above-mentioned values are values before charge-and-discharge initiation, and are fluctuated by charge and discharge.

[0048] According to a desired presentation, grinding mixing of the lithium content multiple oxide used for positive active material by this invention can be carried out, and it can calcinate or compound a carbonate, a nitrate, oxide, or a hydroxide of transition metals, such as the carbonate of a lithium, a nitrate, oxide or a hydroxide, cobalt, manganese, or nickel, etc. by the solution reaction. Especially the calcinating method is desirable and burning temperature is 250-1500 degrees C of the temperature which some mixed compounds decompose and fuse. As for firing time, it is desirable that it is 1 - 80 hours. as a baking gas ambient atmosphere -- air, an oxidizing atmosphere, and reducing atmosphere -- any are especially sufficient and it is not limited.

[0049] In this invention, the positive active material with which plurality differed may be used together. For example, what has the opposite expansion contraction behavior at the time of charge and discharge can be used. The desirable example of the positive active material which expands at the time of discharge (at the time of lithium ion insertion), the desirable example of the positive active material contracted at the time of charge (at the time of lithium ion emission) is a spinel mold lithium content manganic acid ghost, contracts at the time of discharge (at the time of lithium ion insertion), and expands at the time of charge (at the time of lithium ion emission) is lithium content cobalt oxide. As a desirable structure expression of a spinel mold lithium content manganic acid ghost, it is $\text{Li}_{1-x}\text{Mn}_2\text{O}_4$ ($0 \leq x \leq 1$), and is $\text{Li}_{1-x}\text{CoO}_2$ ($0 \leq x \leq 1$) as a desirable example of lithium content cobalt oxide.

[0050] the positive electrode in this invention -- a mixture -- in the constituted cell, if an inner electric conduction agent is an electronic conduction nature ingredient which does not cause a chemical change, it is good anything. For example, organic conductivity ingredients, such as conductive metallic oxide, such as conductive whiskers, such as metal powder, such as conductive fiber, such as carbon black, such as graphite, such as natural graphites (scale-like graphite etc.) and an artificial graphite, acetylene black, KETCHIEN black, channel black, furnace black, lamp black, and thermal black, a carbon fiber, and a metal fiber, fluoride carbon, copper, nickel, aluminum, and silver, a zinc oxide, and potassium titanate, and titanium oxide, or a polyphenylene derivative, etc. can be included as independent or such mixture. In these electric conduction agents, an artificial graphite, acetylene black, and especially nickel powder are desirable. Although especially the addition of an electric conduction agent is not limited, its 1 - 50 % of the weight is especially desirable, and its 1 - 30 % of the weight is desirable. In carbon or especially graphite, 2 - 15 % of the weight is desirable.

[0051] the positive electrode in this invention -- a mixture -- the decomposition temperature of an inner desirable binder is a polymer 300 degrees C or more. For example, polyethylene, polypropylene, polytetrafluoroethylene (PTFE), Polyvinylidene fluoride (PVDF), a tetrafluoroethylene-hexafluoroethylene copolymer, A tetrafluoroethylene-hexafluoropropylene copolymer (FEP), A tetrafluoroethylene-perfluoroalkyl vinyl ether copolymer (PFA), A vinylidene fluoride-

hexafluoropropylene copolymer, a vinylidene fluoride-chlorotrifluoroethylene copolymer, An ethylene-tetrafluoroethylene copolymer (ETFE resin), polychlorotrifluoroethylene resin (PCTFE), A vinylidene fluoride-pentafluoropropylene copolymer, a propylene-tetrafluoroethylene copolymer, An ethylene-chlorotrifluoroethylene copolymer (ECTFE), a vinylidene fluoride-hexafluoropropylene-tetrafluoroethylene copolymer, and a vinylidene fluoride-perfluoromethylvinylether-tetrafluoroethylene copolymer can be mentioned. Especially, this most desirable is polyvinylidene fluoride (PVDF) and polytetrafluoroethylene (PTFE).

[0052] If it is the electronic conductor which does not cause a chemical change in the constituted cell as a charge collector of a positive electrode, it is good anything. For example, the thing which made carbon, nickel, titanium, or silver process is used for the front face of aluminum or stainless steel else [, such as stainless steel, nickel, aluminum, titanium, and carbon] as an ingredient. Especially, aluminum or an aluminium alloy is desirable. Oxidizing the front face of these ingredients is also used. Moreover, irregularity may be attached to a current collection body surface by surface treatment. As for a configuration, the Plastic solid of a film besides the foil, a sheet, the thing netted and punched, a lath object, a porous body, foam, a fiber group, and a nonwoven fabric object etc. is used. Although especially thickness is not limited, a 1-500-micrometer thing is used.

[0053] As a negative-electrode ingredient used by this invention, what is necessary is just occlusion and the compound which can be emitted about lithium ions, such as a lithium, a lithium alloy, an alloy, an intermetallic compound, carbon, an organic compound, an inorganic compound, a metal complex, and an organic high molecular compound. These may be independent, or it may combine and they may use.

[0054] As a lithium alloy, Li-aluminum (U.S. Pat. No. 4002492 etc.), Li-aluminum-Mn, Li-aluminum-Mg, Li-aluminum-Sn, Li-aluminum-In, Li-aluminum-Cd, Li-aluminum-Te, Li-Ga (JP,60-257072,A), Li-Cd, Li-In, Li-Pb, Li-Bi, Li-Mg, etc. are mentioned. In this case, as for the content of a lithium, it is desirable that it is 10% or more.

[0055] As an alloy and an intermetallic compound, the compound of transition metals and silicon, the compound of transition metals and tin, etc. are mentioned, and especially the compound of nickel and silicon is desirable.

[0056] As a carbonaceous ingredient, corks, pyrolytic carbon, a natural graphite, an artificial graphite, a meso carbon micro bead, a graphitization mesophase microsphere, vapor growth carbon, glassy carbon, a carbon fiber (a polyacrylonitrile system, a pitch system, a cellulose system, vapor growth carbon system), indeterminate form carbon, the carbon with which the organic compound was calcinated may be mentioned, these may be independent, or it may combine and they may use. Graphite ingredients, such as what graphitized the mesophase microsphere especially, a natural graphite, and an artificial graphite, are desirable. In addition, different-species compounds, such as O, B, P, N, S, SiC, and B₄C, may also be included in a carbonaceous ingredient besides carbon. As a content, 0 - 10 % of the weight is desirable.

[0057] As an inorganic compound, titanate-acid ghosts, tungstic-acid ghosts, molybdic-acid ghosts, niobic acid ghosts, borazine san ghosts, and ferric acid ghosts are mentioned as for example, a tin compound, a silicon compound, and an inorganic oxide, for example. Moreover, as inorganic chalcogenide, an iron sulfide, a molybdenum sulfide, a titanium sulfide, etc. are mentioned, for example. As an organic high molecular compound, cobalt nitrides, copper nitrides, nickel nitrides, iron nitrides, and manganese nitrides can be used as high molecular compounds, such as the poly thiophene and polyacethylene, and a nitride.

[0058] These negative-electrode ingredients may be compounded and used, for example, the combination of carbon, an alloy and carbon, an inorganic compound, etc. can be considered.

[0059] The average grain size of the carbon material used by this invention has desirable 0.1-60 micrometers. It is 0.5-30 micrometers more preferably. As for specific surface area, it is desirable that it is 1-10m²/g. The magnitude (LC) of the microcrystal of the direction of a c-axis has [a crystal structure top / spacing (d002) of a carbon hexagon-head flat surface] a graphite 100Å or more desirable [moreover,] at 3.35-3.40Å.

[0060] Since Li contains in positive active material in this invention, the negative-electrode ingredients

(carbon etc.) which do not contain Li can be used. Moreover, when the negative-electrode material which does not contain such Li is made to contain a little (for it to be 0.01 - 10 weight section extent to the negative-electrode material 100 weight section) Li, even if a part of Li(s) react with an electrolyte etc. and serve as inactive, since it can supply with Li which the above-mentioned negative-electrode material was made to contain, it is desirable. It is good, in order to make negative-electrode material contain Li as mentioned above, for example, if the lithium metal heated and fused is applied on the charge collector which stuck negative-electrode material by pressure, Li is infiltrated into negative-electrode material, or a lithium metal is beforehand stuck by sticking by pressure etc. into an electrode group and Li is made to dope in a negative-electrode ingredient electrochemically in the electrolytic solution.

[0061] a negative electrode -- a mixture -- an inner electric conduction agent -- a positive electrode -- a mixture -- like an inner electric conduction agent, if it is the electronic conduction nature ingredient which does not cause a chemical change in the constituted cell, it is good anything. Moreover, since the carbonaceous ingredient itself has electronic conduction nature when using a carbonaceous ingredient for a negative-electrode ingredient, even if it contains an electric conduction agent, it is not necessary to carry out.

[0062] a negative electrode -- a mixture -- as an inner binder, although you may be any of thermoplastics and thermosetting resin, in this invention, the decomposition temperature of a desirable binder is a polymer 300 degrees C or more. For example, polyethylene, polypropylene, polytetrafluoroethylene (PTFE), Polyvinylidene fluoride (PVDF), styrene butadiene rubber, a tetrafluoroethylene-hexafluoropropylene copolymer (FEP), A tetrafluoroethylene-perfluoroalkyl vinyl ether copolymer (PFA), A vinylidene fluoride-hexafluoropropylene copolymer, a vinylidene fluoride-chlorotrifluoroethylene copolymer, An ethylene-tetrafluoroethylene copolymer (ETFE resin), polychlorotrifluoroethylene resin (PCTFE), A vinylidene fluoride-pentafluoropropylene copolymer, a propylene-tetrafluoroethylene copolymer, An ethylene-chlorotrifluoroethylene copolymer (ECTFE), a vinylidene fluoride-hexafluoropropylene-tetrafluoroethylene copolymer, and a vinylidene fluoride-perfluoromethylvinylether-tetrafluoroethylene copolymer can be mentioned. They are styrene butadiene rubber and polyvinylidene fluoride more preferably. Styrene butadiene rubber is the most desirable especially.

[0063] If it is the electronic conductor which does not cause a chemical change in the constituted cell as a charge collector of a negative electrode, it is good anything. For example, a thing, an aluminum-Cd alloy, etc. which were made to process carbon, nickel, titanium, or silver are used for the front face of copper or stainless steel else [, such as stainless steel, nickel copper, titanium, and carbon,] as an ingredient. Especially, copper or a copper alloy is desirable. Oxidizing the front face of these ingredients is also used. Moreover, irregularity may be attached to a current collection body surface by surface treatment. As for a configuration, a film besides the foil, a sheet, the thing netted and punched, a lath object, a porous body, foam, the Plastic solid of a fiber group, etc. are used. Although especially thickness is not limited, a 1-500-micrometer thing is used.

[0064] an electrode -- a filler besides an electric conduction agent or a binder, a dispersant, an ion electric conduction agent, a pressure enhancement agent, and other various additives can be used for a mixture. In the constituted cell, a filler can be used anything, if it is the fibrous ingredient which does not cause a chemical change. Usually, fiber, such as olefin system polymers, such as polypropylene and polyethylene, glass, and carbon, is used. Although especially the addition of a filler is not limited, 0 - 30 % of the weight is desirable.

[0065] The positive electrode and negative electrode in this invention may have the protective layer introduced for the purpose of the under coat introduced for the purpose other than the binder layer containing positive active material or a negative-electrode ingredient, such as amelioration of adhesion and conductivity of a charge collector and a binder layer, a cycle property, and charge-and-discharge effectiveness, the mechanical protection of a binder layer, or chemical protection. This under coat and protective layer can contain a binder, an electric conduction agent particle, a particle without conductivity, etc.

[0066] The nonaqueous electrolyte in this invention consists of a solvent and lithium salt which dissolves in the solvent. As a non-aqueous solvent, for example Ethylene carbonate (EC), propylene carbonate (PC), Annular carbonate, such as butylene carbonate (BC) and vinylene carbonate (VC) Dimethyl carbonate (DMC), diethyl carbonate (DEC), Un-annular carbonate, such as ethyl methyl carbonate (EMC) and dipropyl carbonate (DPC) Aliphatic series carboxylate, such as methyl formate, methyl acetate, methyl propionate, and ethyl propionate gamma-lactone, such as gamma-butyrolactone, 1, 2-dimethoxyethane (DME), Non-cyclic ether, such as 1, 2-diethoxy ethane (DEE), and ethoxy methoxyethane (EME) Cyclic ether, such as a tetrahydrofuran and 2-methyl tetrahydrofuran Dimethyl sulfoxide, 1, 3-dioxolane, a formamide, an acetamide, Dimethylformamide, dioxolane, an acetonitrile, propyl nitril, Nitromethane, ethyl mono-glyme, trialkyl phosphate, trimethoxy methane, A dioxolane derivative, a sulfolane, a methyl sulfolane, 1,3-dimethyl-2-imidazolidinone, 3-methyl-2-oxazolidinone, a propylene carbonate derivative, Aprotic organic solvents, such as a tetrahydrofuran derivative, ethyl ether, 1, 3-propane ape ton, an anisole, dimethyl sulfoxide, and N-methyl pyrrolidone, can be mentioned, and these kinds or two sorts or more are mixed and used. It is desirable to use mixed stock with the mixed stock of annular carbonate and un-annular carbonate or annular carbonate and un-annular carbonate, and aliphatic series carboxylate as a principal component especially.

[0067] As lithium salt which dissolves in these solvents For example, LiClO_4 , LiBF_4 , LiPF_6 , LiAlCl_4 , LiSbF_6 , LiSCN , LiCl , LiCF_3SO_3 , LiCF_3CO_2 , $\text{Li}(\text{CF}_3\text{SO}_2)_2$, LiAsF_6 and $\text{LiN}(\text{CF}_3\text{SO}_2)_2$, $\text{LiB}_{10}\text{Cl}_{10}$ (JP,57-74974,A), A low-grade aliphatic-carboxylic-acid lithium (JP,60-41773,A), LiCl , LiBr , LiI (JP,60-247265,A), Although it can be used for the electrolytic solution which can mention a chloro borane lithium (JP,61-165957,A), 4 phenyl lithium borate (JP,61-214376,A), etc., and uses these combining independent or two sorts or more It is more desirable to include especially LiPF_6 .

[0068] Especially the desirable nonaqueous electrolyte in this invention is the electrolytic solution which contains LiPF_6 as lithium salt, including ethylene carbonate and ethyl methyl carbonate at least. although especially the amount that adds these electrolytic solutions in a cell is not limited -- positive active material, the amount of a negative-electrode ingredient, and the size of a cell -- **** for initial complements -- things are made. Although especially the amount of dissolutions to the non-aqueous solvent of lithium salt is not limited, its 0.2 - 2 mol/l is desirable. It is more desirable to consider as 0.5 - 1.5 mol/l especially.

[0069] Moreover, if needed, organic additives, such as 2-methyl furan, a thiophene (JP,61-161673,A), a pyrrole (JP,3-59963,A), an aniline (JP,60-79677,A), crown ether, a pyridine, a triethyl FOSU fight, triethanolamine, cyclic ether, ethylenediamine, n-glyme, hexalin acid TORIAMIDO, a nitrobenzene derivative, and a nitrogen-containing aromatic heterocycle compound (JP,9-204932,A), may be dissolved in the above-mentioned electrolytic solution in order to acquire a good charge-and-discharge property. It is usually used for them, making separators, such as a porous polymer, a glass filter, and a nonwoven fabric, sink in or fill up with this electrolytic solution.

[0070] Moreover, in order to make the electrolytic solution into incombustibility, a halogen-containing solvent, for example, a carbon tetrachloride, and a 3 fluoridation ethylene chloride can be included in the electrolytic solution. Moreover, in order to give fitness to elevated-temperature preservation, carbon dioxide gas can be included in the electrolytic solution.

[0071] Moreover, the gel electrolyte which made the organic solid electrolyte contain the above-mentioned nonaqueous electrolyte can also be used. The above-mentioned organic solid electrolyte has effective macromolecule matrix materials, such as polyethylene oxide, polypropylene oxide, polyphosphazene, the poly aziridine, a polyethylene sulfide, polyvinyl alcohol, polyvinylidene fluoride, poly hexafluoropropylene, etc. these derivatives, mixture, and complex. Especially, the mixture of vinylidene fluoride, the copolymer of hexafluoropropylene and polyvinylidene fluoride, and polyethylene oxide is desirable.

[0072] As a separator, it has big ion transmittance, and has a predetermined mechanical strength, and an insulating fine porosity thin film is used. Moreover, it is desirable to have the function to blockade a hole above 80 degrees C and to raise resistance. Independent, or the sheets and nonwoven fabrics which were built with a combined olefin system polymer or a glass fiber, such as polypropylene and

11-13th

polyethylene, are used from organic solvent-proof nature and hydrophobicity. As for the aperture of a separator, it is desirable that it is the range which the active material from which it was desorbed from the electrode sheet, a binder, and an electric conduction agent do not penetrate, for example, what is 0.01-1 micrometer is desirable. Generally as for the thickness of a separator, 10-300 micrometers is used. Moreover, although a void content is determined according to the permeability, the material, and film pressure of an electron or ion, it is desirable that it is generally 30 - 80%.

[0073] The configuration of a cell can apply a coin mold, a carbon button mold, a sheet mold, cylindrical, a flat mold, a square shape, etc. to all. The configuration of a cell is compressed into the configuration of a pellet at the time of a coin mold or a carbon button mold, and the mixture of positive active material or a negative-electrode ingredient is mainly used. The thickness and the diameter of the pellet are decided with the magnitude of a cell. Moreover, when the configurations of a cell are a sheet mold, cylindrical, and a square shape, on a charge collector, the mixture of positive active material or a negative-electrode ingredient is applied, (coat) dried and compressed, and is mainly used. A general approach can be used for the method of application. For example, the reverse roll method, the direct rolling method, the blade method, the knife method, the extrusion method, the curtain method, the gravure method, the bar method, the casting method, a dip method, and the squeeze method can be mentioned. The blade method, the knife method, and the extrusion method are desirable also in it. As for spreading, it is desirable to carry out the rate for 0.1-100m/. Under the present circumstances, according to the solution physical properties of a mixture, and drying, the surface state of a good spreading layer can be acquired by selecting the above-mentioned method of application. The time of one side [every] ** or double-sided coincidence is sufficient as spreading. Moreover, it is desirable to prepare a spreading layer in the both sides of a charge collector, and you may consist of two or more layers in which the spreading layer of one field contains a binder layer. A binder layer contains a binder, an electrical conducting material, etc. other than the matter concerning insertion emission of a lithium ion like positive active material or a negative-electrode ingredient. You may have the protective layer which does not contain the active material other than a binder layer, the under coat prepared on a charge collector, the interlayer prepared between binder layers. As for the layer which does not have these active materials, it is desirable that a conductive particle, an insulating particle, and a binder are included.

[0074] Moreover, continuation, an intermission, or a stripe is sufficient as spreading. Although the thickness, die length, and width of the spreading layer are decided with the magnitude of a cell, the thickness of the spreading layer of one side is in the condition of having been compressed after dry, and especially its 1-2000 micrometers are desirable.

[0075] The approach generally adopted can be used as a pellet, or desiccation of a sheet or the dehydration approach. Especially, it is desirable independent or to combine and to use hot blast, a vacuum, infrared radiation, far infrared rays, an electron ray, and a damp wind. The range of temperature of 80-350 degrees C is desirable, and it is especially desirable. [of the range which is 100-250 degrees C] moisture content -- the cell whole -- 2000 ppm or less -- desirable -- a positive electrode -- a mixture and a negative electrode -- it is desirable to make it 500 ppm or less in a mixture or an electrolyte, respectively in respect of cycle nature. Although the approach generally adopted can be used for the method of pressing a sheet, a die-press method and its calender pressing method are especially desirable. Although especially press ** is not limited, its 0.2 - 3 t/cm² is desirable. The pressing speed of the calender pressing method has the amount of desirable 0.1-50m/. Press temperature has desirable room temperature -200 degree C. As for the ratio of the width of face of the positive-electrode sheet to a negative-electrode sheet, 0.9-1.1 are desirable. Especially, 0.95-1.0 are desirable. the content ratio of positive active material and a negative-electrode ingredient -- a compound class and a mixture -- although it cannot limit since it changes with formulas, it can be set as the optimal value in the viewpoint of capacity, cycle nature, and safety.

[0076] In addition, the winding objects of the electrode in this invention may be the prismatic form configurations where it is not necessary to be necessarily a perfect circle cartridge, and the cross section is an ellipse, such as an ellipse cartridge and a rectangle.

[0077] Although it is desirable to put the desirable thing of the above-mentioned chemistry ingredient or a cell component part together as for a desirable combination of this invention, both acetylene black is also included as an electric conduction agent especially as positive active material including Li_xCoO_2 , Li_xNiO_2 , and $\text{Li}_x\text{Mn}_2\text{O}_4$ (it is $0 \leq x \leq 1$ here). The positive-electrode charge collector is carrying out configurations, such as the network and sheet which are made from stainless steel or aluminum, a foil, and a lath. As a negative-electrode ingredient, it is not lithium metal independent, and it is desirable that at least one sort of compounds, such as an alloy and a carbonaceous ingredient, are included. The negative-electrode charge collector is carrying out configurations, such as the network and sheet which are made from stainless steel or copper, a foil, and a lath. In the mixture used with positive active material or a negative-electrode ingredient, carbon materials, such as acetylene black and a graphite, may be mixed as an electronic conduction agent. a binder is independent in elastomers, such as a polymer containing fluorine-containing thermoplasticity compounds, such as polyvinylidene fluoride and polytetrafluoroethylene, and an acrylic acid, styrene butadiene rubber, and ethylene-propylene terpolymer, -- or it can mix and use. Moreover, as the electrolytic solution, it combines and ethylene carbonate and the thing containing LiPF_6 which added aliphatic series carboxylate compounds, such as methyl acetate and methyl propionate, to annular [, such as diethyl carbonate, dimethyl carbonate, and ethyl methyl carbonate,], un-annular carbonate, or them are still more desirable as lithium salt. Furthermore, independent or those combination of polypropylene or polyethylene are desirable as a separator. Any of a cylinder, flatness, and a square shape are sufficient as the gestalt of a cell. It is desirable to equip a cell with a means (an example, an internal pressure open sand mold relief valve, separator that raises resistance at an elevated temperature) by which insurance is securable also for malfunction.

[0078]

[Example] Hereafter, the example of this invention is explained, referring to a drawing.

[0079] (Example 1) Drawing of longitudinal section of the cylindrical shape cell used for drawing 1 by this example is shown. In drawing, the cell case into which 1 processed the stainless steel plate of organic-proof electrolytic-solution nature, the obturation plate with which 2 prepared the relief valve, and 3 show insulating packing. 4 is a group of electrode, and a positive electrode and a negative electrode are wound in the shape of a multiple-times swirl through a separator, and it is contained in the case 1. And from the above-mentioned positive electrode, the positive-electrode lead 5 is pulled out and it connects with the obturation plate 2, and from the negative electrode, the negative-electrode lead 6 is pulled out and it connects with the pars basilaris ossis occipitalis of the cell case 1. 7 is prepared in the vertical section of a group of electrode 4 with the insulating ring, respectively. The forward one, a negative-electrode plate, etc. are explained in detail below.

[0080] a positive electrode mixes Li_2CO_3 and Co_3O_4 , mixes 7% of fluororesin system binders acetylene black 3% to the weight of the powder of LiCoO_2 which calcinated for 10 hours and was compounded at 900 degrees C, and suspends them in a carboxymethyl-cellulose water solution -- making -- a positive electrode -- a mixture -- it considered as the paste. aluminum foil with a thickness of 30 micrometers -- a positive electrode -- a mixture -- coating of the paste was carried out, and it rolled out after desiccation and considered as the positive-electrode plate with the thickness of 0.18mm, a width of face [of 37mm], and a die length of 390mm.

[0081] The negative electrode used what graphitized the mesophase microsphere at the 2800-degree C elevated temperature (a mesophase graphite is called below). After mixing styrene / butadiene rubber 5% to the weight of this mesophase graphite, the carboxymethyl-cellulose water solution was made to suspend and it was made the shape of a paste. and this negative electrode -- a mixture -- coating of the paste was carried out to both sides of Cu foil with a thickness of 0.02mm, and it rolled out after desiccation and considered as the negative-electrode plate with the thickness of 0.20mm, a width of face [of 39mm], and a die length of 420mm.

[0082] And the lead made from nickel was attached in the product made from aluminum, and the negative-electrode plate at the positive-electrode plate, respectively, and the cell case with winding, a diameter [of 17.0mm], and a height of 50.0mm was spirally supplied through the separator made from

polypropylene with the thickness of 0.025mm, a width of face [of 45mm], and a die length of 950mm. What dissolved LiPF₆ of 1 mol/l in the solvent which mixed MP with EC by the volume ratio of DEC and 30:50:20 is used for the electrolytic solution. As an additive of the electrolytic solution As an organic compound applicable to (** 1), hexa phenylbenzene, para terphenyl, 1-phenyl piperazine, 1, 2 and 3, 4-tetrahydroisoquinoline, Phenylcyclohexane, 1 and 3, 5-triphenyl benzene, dodeca hydronalium triphenylene, and a divinylbenzene were added 2% of the weight to the total amount of an organic solvent and an additive, after pouring this in, it obturated, and it considered as the cells 1-9 of this invention.

[0083] (Example 2) The telescopic cell of the eddy coil former was constituted like (the example 1) except having used t-butyl vinyl ether, a methyl-methacrylate monomer, a vinyl cyclohexane, and 2,3-dimethyl-1,3-butadiene as an organic compound applicable to (** 2) as an additive of the electrolytic solution. This was made into the cells 10-14 of this invention.

[0084] (Example 3) The telescopic cell of the eddy coil former was constituted like (the example 1) except having used tribenzylamine and N-phenyl-dibenzylamine as an organic compound applicable to (** 3) as an additive of the electrolytic solution. This was made into the cells 15 and 16 of this invention.

[0085] (Example 4) The telescopic cell of the eddy coil former was constituted like (the example 1) except having used squalene or (E)-beta-farnesene as an organic compound applicable to the straight chain mold organic compound of the carbon numbers 15-60 which have a double bond as an additive of the electrolytic solution. This was made into the cells 15 and 16 of this invention.

[0086] (Example 5) The telescopic cell of the eddy coil former was constituted like (the example 1) except having used a tris (4-methoxypheny) phosphine, a (1R)-(+)-alpha pinene, a dicyclopentadiene, the II (ethylene glycol) divinyl ether, 9, 10-dihydroanthracene, TORIPUCHISEN, and [2, 2] PARASHI clo fan as an additive of the electrolytic solution. This was made into the cells 19-25 of this invention.

[0087] (Example 1 of a comparison) As an example of a comparison, the telescopic cell of the eddy coil former was constituted for the cell which does not add the additive of the electrolytic solution like (the example 1). It considered as the cell (cell 26) of a comparison of this.

[0088] (Example 2 of a comparison) As an example of a comparison, the telescopic cell of the eddy coil former was constituted like (the example 1) except having used the biphenyl, the thiophene, and 3-chloro thiophene as an additive of the electrolytic solution. It considered as the cell (cells 27-29) of a comparison of this.

[0089] The cells 1-25 of this invention and five every cels each of comparative cells 26-29 are prepared. Next, with the environmental temperature of 20 degrees C After investigating the discharge property in 1A of the cell of a charge condition which performed constant-potential charge of 500mA of charge electrical-potential-difference 4.2V, and charging-time 2 hours of limit currents, It saves for five days at 80 degrees C in the state of charge, and the result of having performed charge and discharge on conditions with the same said of the cell after preservation, and having asked for the capacity recovery factor after preservation (capacity x100(%) before the capacity/preservation after preservation) is shown in Table 1 - (Table 6).

[0090] Moreover, these were prepared 20 every cels each, and it overcharged from the charge condition by 1A to the pan, and checked whether a cell would carry out abnormality generation of heat. (Table 1) The number of cels which abnormality generation of heat in 20 cels generated in - (Table 6) was shown.

[0091]

[Table 1]

(実施例 1)

No.	添加剤	HOMO	LUMO	保存後回復率	過充電試験
1	ヘキサフェニルベンゼン	-9.442	0.0813	91.2	0/20
2	p-テルフェニル	-9.3829	0.0474	93.7	0/20
3	1-フェニルピペラジン	-8.8402	0.3045	94.8	0/20
4	1,2,3,4-テトラヒドロイソキノリン	-9.1063	0.3747	92.8	0/20
5	フェニルシクロヘキサン	-9.4288	0.3961	92.3	0/20
6	1,3,5-トリフェニルベンゼン	-9.5698	0.1704	93.7	0/20
7	ドデカヒドロトリフェニレン	-8.8282	0.5063	91.4	0/20
8	ジビニルベンゼン	-9.0738	-0.1222	91.7	0/20
9	1,4-ジシクロヘキシルベンゼン	-9.2999	0.3508	91.7	0/20

[0092]

[Table 2]

(実施例 2)

No.	添加剤	HOMO	LUMO	保存後回復率	過充電試験
10	トブチルビニルエーテル	-9.3128	1.4402	93.5	0/20
11	メタクリル酸メチルモノマー	-10.56	0.4128	91.5	0/20
12	ビニルシクロヘキサン	-10.124	1.1991	91.7	0/20
13	2,3-ジメチル-1,3-ブタジエン	-9.5072	0.6255	91.5	0/20
14	メチレンシクロヘキサン	-9.8256	1.1529	94.6	0/20

[0093]

[Table 3]

(実施例 3)

No.	添加剤	HOMO	LUMO	保存後回復率	過充電試験
15	トリベンジルアミン	-9.0909	0.1285	94.8	0/20
16	N-フェニルジベンジルアミン	-9.2571	0.2468	92.6	0/20

[0094]

[Table 4]

(実施例 4)

No.	添加剤	HOMO	LUMO	保存後回復率	過充電試験
17	スクアレン	-9.2002	0.9853	93.4	0/20
18	(E)- β -ファルネセン	-9.3018	0.3382	91.5	0/20

[0095]

[Table 5]

(実施例 5)

No.	添加剤	HOMO	LUMO	保存後回復率	過充電試験
19	トリス(4-メトキシフェニル)ホスフィン	-8.7072	0.1294	94.1	0/20
20	(1R)-(+)- α -ピネン	-9.6117	1.1448	94.3	0/20
21	ジシクロペンタジエン	-9.5743	0.9682	93.2	0/20
22	ジ(エチレングリコール)ジビニルエーテル	-9.5991	1.1676	92.5	0/20
23	8,10-ジヒドロアントラセン	-9.171	0.3121	91.7	0/20
24	トリブチセン	-9.2378	0.1824	93.9	0/20
25	[2.2]ナラシクロファン	-8.9716	0.2534	91.1	0/20

[0096]

[Table 6]

(比較例 1, 2)

No.	添加剤	HOMO	LUMO	保存後回復率	過充電試験
26	なし			93.9	8/20
27	ビフェニル	-9.1449	-0.1419	78.4	0/20
28	チオフェン	-9.5429	-0.1917	72.3	0/20
29	3-クロロチオフェン	-9.2515	-0.4247	69.6	0/20

[0097] As shown in (Table 6), when it overcharged, about the cell 26 which does not add an additive, the phenomenon of abnormality generation of heat was accepted in 8 in 20 cels cel. Although the safety of a cell has been secured in the former by establishing two or more safety devices depended on the protection network and current cutoff device of a charge electrical potential difference, when not performing safety protection, for example like this example 1 of a comparison, such abnormality generation of heat may take place.

[0098] The result of having measured the electrical potential difference and cell side-face temperature at the time of overcharge of the example 1 (cell 26) of a comparison to drawing 2 is shown.

[0099] By overcharging shows that the lithium in a positive electrode was drawn out first, cell temperature rose after that based on increase of electronic resistance of a positive electrode, increase of the liquid resistance by the oxidative degradation of the electrolytic solution, an exhaustion of liquid, etc., and abnormality generation of heat has arisen since thermal stability has already fallen in the melting temperature of a separator.

[0100] On the other hand, by the cells 1-25 which added the additive of this invention, as shown in Table 1 - (Table 5), even if it performed the overcharge trial, abnormality generation of heat was not accepted. As an example of representation of this example, the overcharge behavior of the cell which added the hexa phenylbenzene of an example 1 was shown in drawing 3.

[0101] In this example, it turns out that cell voltage will rise remarkably shortly after overcharge is started, and the polarization property of a cell is getting worse. In connection with this, cell temperature also rises early, a current will not flow in the melting temperature of a separator, and temperature falls to insurance, without cell temperature carrying out abnormality generation of heat.

[0102] This is for a current not to flow on a cell by an exhaustion of the electrolytic solution, or the shutdown of a separator, before an additive generates the high oxidation-polymerization film of resistance on a positive-electrode front face according to an oxidation polymerization preferentially, disassembly of the electrolytic solution advances dominantly when polarization becomes large and the

thermal stability of a positive electrode falls rather than the lithium in a positive electrode is drawn out in overcharge.

[0103] Dependability can be further raised as a cell charge system by preparing a positive thermistor (PTC) or a thermal fuse as a function to sense the rise of cell temperature and to cut the circuit of charge naturally. Since it is indispensable for positive active material to operate at stable temperature thermally, and not to operate in the general environmental temperature of a cell device as for the operating temperature of a positive thermistor (PTC) or a thermal fuse, it is desirable that it is the range which is 60-120 degrees C.

[0104] Since the additive used by this example was excellent in the reducibility-proof in a negative electrode, compared with the additive advocated conventionally used in the example 2 of a comparison, the preservation property in an elevated temperature was excellent.

[0105] As mentioned above, by using the additive of this invention showed that a very reliable cell with a good elevated-temperature preservation property was realizable safely at the time of overcharge.

[0106] Moreover, as a result of performing examination to the concentration of an additive, improvement in safety was accepted at 0.1 % of the weight or more. The discharge property of a cell began to worsen [an addition] at 20 % of the weight or more. Since own conductivity of the electrolytic solution decreased, this is considered.

[0107] As for the addition of the additive from this to the electrolytic solution, it is desirable that it is 0.1 - 20% of the weight of the range.

[0108] In addition, this invention is not limited to this example.

[0109]

[Effect of the Invention] It is the nonaqueous electrolyte rechargeable battery characterized by adding an electrolytic-solution additive in 0.1 - 20% of the weight of the range to the sum total of the solvent in the electrolytic solution, and an additive in this invention as mentioned above. By adding the additive of this invention, it is safe also at the time of overcharge, and a cell with the high dependability excellent in the preservation property can be realized. As a separator, if the fine porosity polyolefine film is used, dependability of this cell will improve further.

[0110] Moreover, the charge control system of a still more reliable nonaqueous electrolyte rechargeable battery and a nonaqueous electrolyte rechargeable battery can be offered by giving the function to sense the rise of cell temperature and to cut the circuit of charge.

[0111] By using the charge control system of such a nonaqueous electrolyte rechargeable battery and a nonaqueous electrolyte rechargeable battery, safety is high and devices, such as the cellular phone which was excellent in the life property in the long run, a Personal Digital Assistant device, a camcorder, a personal computer, PDA, a pocket audio equipment, an electric vehicle, and a power source for load leveling, can be offered.

[Translation done.]